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HIGH INTENSITY LIGHT PROPAGATION IN InAs (Preprint)

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14. ABSTRACT

We present our experimental and theoretical results on nonlinear absorption of light in InAs. The nonlinear variation of output intensity as a function of input intensity and time are calculated by solving four coupled rate equations simultaneously. All required quantities, including two-photon absorption, free-carrier absorption, Auger and radiative recombination lifetimes, and intrinsic carrier densities, have been obtained from the underlying band structures. The calculated thickness and energy-dependent output intensities in InAs agree very well with the values measured in our pump-probe experiments.

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High Intensity Light Propagation in InAs

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Abstract

We present our experimental and theoretical results on nonlinear absorption of light in InAs. The nonlinear variation of output intensity as a function of input intensity and time are calculated by solving four coupled rate equations simultaneously. All required quantities, including two-photon absorption, free-carrier absorption, Auger and radiative recombination lifetimes, and intrinsic carrier densities, have been obtained from the underlying bandstructures. The calculated thickness and energy-dependent output intensities in InAs agree very well with the values measured in our pump-probe experiments.

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Nonlinear absorption in semiconductors continues to be of scientific and technological interest.[1–4] Technologically, nonlinear absorption (NLA) is being used in a number of applications, including frequency conversion, optical switches and optical limiting.[1] The tmportance of free carrier effects in controlling the infrared nonlinear optical properties has long been realized[2–4]. Consequently, considerable efforts were undertaken to understand the NLA and evaluate the parameters such as two-photon absorption (TPA) coefficient, free-carrier absorption (FCA) coefficient, refraction cross section, and lifetimes in semiconductors.[5–10] However, extracting parameters from the measured values usually requires careful design of experiments to isolate and remove the effect of other parameters on the observed results. Even when evaluated at appropriate limits, the extraction ignores the complicated interdependence of these parameters on temperature and carrier density. For example, TPA and FCA depend on carrier density, which in turn depends on temperature. Consequently, the extracted values differ substantially from each other.[4–6,9,10] In addition, it is not clear whether the values extracted in certain limits can be used in modeling of high-intensity light propagation where the photoexited carrier distribution is far from equilibrium. There have also been several intuitive calculations to evaluate these parameters using either effective mass bands and **k.p** bands with adjustable matrix elements.[3,7–10] The trends are often obtained correctly. However, for complete understanding and modeling of light propagation with improved predictability, an accurate evaluation of these parameters from full bandstructures needed to be carried out.

In this Letter, we report the results of transmission measurements carried out with (a)

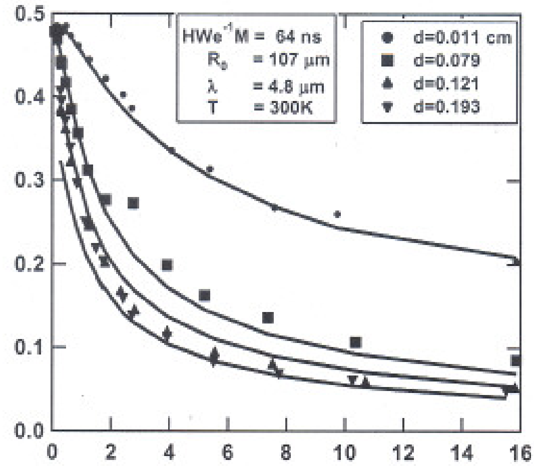

FIG. 1: Comparison of transmitted pump intensity measured (data points) at 300 K in 10^{15} cm^{-3} n-doped InAs with that calculated (solid lines) for four thicknesses.

pump only and (b) the pump-probe technique on a well-characterized InAs sample along with rigorous calculation of time (t) and space (z) dependent transmitted pump and probe intensity. All physical quantities including TPA and FCA coefficients, Auger recombination (AR) and radiative recombination (RR) lifetimes are calculated as functions of temperature (T) and carrier density (N) using full bandstructures. The calculated value of the transmitted probe intensity at the exit surface explains the experimentally observed trend very well and an excellent quantitative agreement is obtained when a Shockley-Read-Hall (SRH) mechanism is also included.

In our pump-probe experiment, the charge carriers were generated using TPA. In addition to the convenient availability of below band gap lasers, the TPA used here ensures that the entire thickness of the sample is utilized for carrier generation at low incident irradiance. The 4.8 μm pump source was obtained by frequency doubling a TEA CO_2 laser to provide 128 ns (full-width at e^{-1} of the maximum) duration pulses with energies up to 10 mJ. First, we carried out a set of NLA experiments at room temperature with only the pump present.

In Figure 1, the intensity transmission of the sample is plotted as a function of incident intensity for four different InAs sample thicknesses (d= 0.11 mm, 0.79 mm, 1.21 mm, and 1.93 mm). With increasing intensity, more carriers are created by TPA, and the resulting FCA decreases the output intensity nonlinearly with input intensity. As the thickness of the sample is increased, the path length of the light is longer and consequently more NLA takes

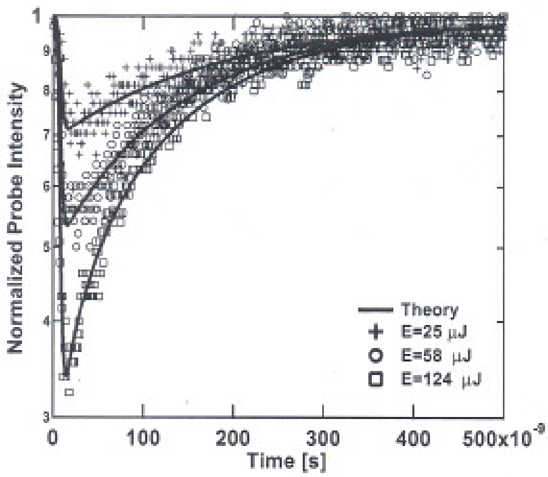

FIG. 2: Comparison of transmission intensity measured (data points) at 300 K in 10^{15} cm^{-3} n-doped InAs with that calculated (solid lines) for three beam energies.

place, as shown in Figure 1.

The decay time of the TPA excited carriers is studied using a pump-probe method. The pump is the 4.8 μm beam described above and the probe is the *cw* laser beam obtained by a grating tuned CO laser operating at 5.3 μm with power of 100 mW. The probe beam was chopped by a mechanical shutter to a 5 ms duration pulse. A dichroic filter (coated for high reflection at 4.8 μm and anti-reflection at 5.3 μm for 45 degree angle of incidence) was used to combine the pump and probe beams which were focused on the sample with the same 200 mm focal length lens to spot sizes of 378 μm and 166 μm, respectively (beam radius at e^{-1} of maximum). The sample studied is a 1-mm-thick InAs wafer with anti-reflection coating. The probe transmission was measured using a HgZnCdTe detector with a rise time less than 1 ns, and the incident and transmitted pump energies were measured using pyroelectric detectors.

Figure 2 displays the time dependence of the transmitted probe intensity (data points) through the sample for three different pump beam energies. At the onset of the pump pulse (t=0), TPA in the material generates electron-hole pairs. Thus created free carriers absorb probe beam, by FCA, and cause a decrease in probe transmission. The transmission is minimum at the peak of the pump pulse. As the free carriers recombine, their density decreases and the FCA decreases. Consequently, the probe transmission increases to eventually reach its initial value. With an increase in the pump energy, from 25 μJ to 124 μJ, more carriers

are created at the end of the pulse and, consequently, the minimum in probe transmission is reduced further.

For a detailed understanding of the mechanisms affecting the light propagation in the material, a complete solution to the following rate equations is needed.

$$\frac{dI_P}{dz} = -\alpha I_P - \beta I_P^2 - \sigma(N_0 + N)I_P, \tag{1}$$

$$\frac{dN}{dt} = \frac{\beta I_P^2}{2h\nu} - \frac{N}{\tau_R} \tag{2}$$

$$\frac{dT}{dt} = \frac{\sigma^*(N_0 + N)I_P}{c_v} + \frac{\beta I_P^2}{c_v} \tag{3}$$

$$\frac{dI_{pr}}{dz} = -\sigma(N_0 + N)I_{Pr}, \tag{4}$$

The equations relate the pump and probe intensities, I_P and I_{pr}, the concentration of the generated charge carriers N, and T. α, and β are the one- and two- photon absorption coefficients, respectively, and σ (σ^*) denote the sum of (difference in) the cross sections for FCA arising from photon emission and absorption. The total recombination rate τ_R^{-1} is the sum of the AR rate (τ_{ar}^{-1}), RR rate (τ_{rr}^{-1}), and SRH rate (τ_{srh}^{-1}). N_0 is the intrinsic carrier density at a given T. We have considered T increase due to the FCA and TPA, which has not been included in previous studies. We find that the increase in T can be significant (as much as 150 K for moderate intensites). It is important to realize that N_0, α, β, σ, and τ_R are T-dependent. In addition, β, τ_R are dependent on N, which in turn depends on T. Moreover, the carrier density created by the pump beam affects the absorption of the probe beam. Hence, Eq. 1 through 4 have to be solved self-consistently. For the chosen value of incident pump intensity I_P(t=0,r,z=0) at a radial distance r, the time-dependent probe intensity I_{pr}(t,r,z) is evaluated at the exit side (z=L) of the sample to compare with experiments. Since the photon energy, hν, is smaller than the bandgap of the materials considered, α is zero. The self-consistent solution requires that N_0, β, σ, and τ_R be calculated at all T and N values of interest. We calculate them in the range – 10^{15} cm^{-3} $\leq$ N $\leq$ 10^{18} cm^{-3} and 80 K $\leq$ T $\leq$ 500K.

The TPA and FCA coefficients, RR and AR lifetimes, Fermi functions and intrinsic carrier concentrations are calculated using a hybrid pseudopotential tight-binding Hamiltonian that includes all long-range interactions.[11] In general, the β is assumed to be N and T dependent. However, we see that the Moss-Bernstein effect offers serious limitations to β at moderate

concentrations and low temperatures. For a given value of (equal electron and hole density) N and T, we first obtain the quasi-Fermi level, as appropriate for the non-equilibrium studies considered here. Then, using appropriate electron and hole distributions (for final and initial states, respectively), β is calculated.[12] We find that β (at 4.8 μm) remains nearly constant (0.45 cm/MW for InAs) for all N until approximately $10^{17}\mathrm{cm}^{-3}$ is reached, after which it decreases like a Fermi function for the given T and could be fitted very well to a functional form of $\beta = a + be^{-c(T)n}$, where n is carrier density in the unit of 10^{-15} cm^{-3}. Of the parameters (a,b,c), we found only c depends on T. N_0, which is normally assumed to be constant,[8] is found to change by two orders of magnitude when the T is changed from 80K to 500K.

The FCA coefficient evaluated with full bands[12] is found to be proportional to N, in agreement with that assumed in the literature.[8] However, the proportionality constant, σ, which is the FCA cross section, is found to be strongly T-dependent. The contribution to the FCA comes more from holes than from electrons. The transition from light-hole to heavy-hole band does not require phonons, whereas the absorption of light by electrons is possible only with phonons. The T-dependence of phonon and electron distribution results in strongly T-dependent FCA cross section.

We modified the previously developed[13] full-bandstructure calculation of the lifetimes associated with electron-hole recombination through Auger and radiative processes to address the issue of electron and hole quasi Fermi levels. The calculated values of the lifetimes could be fitted accurately to a function form, $\tau \equiv G(T)N^{-p(T)}$.

All T-dependent parameters (N,c,G,p,σ) are fitted accurately to a 4th order polynomial in T, for easier and more efficient use in obtaining self-consistent solutions to rate equations. The details of carrier density, coefficient, and recombination rate variation with T and N, and the fitted parameters, will be given elsewhere. Here we present only the transmission results obtained using these calculated values and parameters.

The calculated values of β(N,T), $\sigma(T)$, and τ_R(N,T) are substituted in Eqs. 1-4 and solved for I_{pr}(z,r,t) and N(z,r,t) by the finite difference method with the boundary conditions $I_P(0,r,t) = I_P^0 e^{-(\frac{t-t_0}{\tau_0})^2} e^{-(\frac{r}{r_P})^2}$, $N(z,r,0) = 0$, $T(z,r,0) = T_{Latt}$, and $I_{pr}(0,r,t) = I_{pr}^0 e^{-(\frac{r}{r_{pr}})^2}$, where r_P and r_{pr} are the pump and probe radii. The beam is assumed to be radially symmetric, and the beam energy, E, is the time and r-integrated value of I_P(0,r,t). Care is taken to yield a numerically stable solution even under illumination of a strong pump laser

pulse. For a given incident pump irradiance, we obtain the value of the probe transmission as a function of r, t and z in the sample. Then the transmission intensity is integrated over r to obtain a value at a given z. The transmission intensity evaluated at the exit surface (z=L) is compared with experimental transmission curve. Although the calculated curve has trends similar to that observed, we find that inclusion of the SRH mechanism is needed for quantitative comparison with experiment. The SRH scattering is not intrinsic to the material and depends on external variables such as growth temperature and pressure. Our detailed calculations carried out previously[14] indicate a nearly T-independent SRH lifetime of 200 ns in 2×10^{16} cm^{-3} doped InAs. Although the InAs sample studied here is only 10^{15} cm^{-3} n-doped, the excess carriers generated by the pump is of the order of 1-2 $\times$ 10^{16} cm^{-3}, and justifies the use of the above calculated SRH lifetimes. We further assume that SRH lifetime decreases linearly with excess carrier density and obtain $I_{pr}(L,t)$ in InAs for three values of E. The substrate ambient temperature 300K is used.

First, the equations are solved only for transmitted pump intensity without any probe present. We see in Fig. 1 that the calculated values (solid lines) as a function of input intensity agree very well with the values measured for four different InAs thicknesses. Then, for a fixed InAs thickness and three different beam energies, the transmitted probe intensity is calculated and compared with experiment, as shown in Fig. 2. The calculations explain the observed variations very well. It is important to note that good agreement is obtained for the pump transmission, probe transmission recovery, and the minimum of the probe transmission, which depend critically on all quantities: the intrinsic carrier density, β, σ, temperature and the lifetimes. Only with N-dependence of SRH, included here in an approximation, an excellent agreement with experiment is obtained. The slight disagreement at large t is possibly because of actual N-dependence of SRH lifetimes, not considered here.

In conclusion, we have used pump-probe experiments to understand the optical interaction in InAs. The results are explained with accurately calculated TPA, FCA, AR, and RR lifetimes, and iterative solutions to coupled rate equations. Our calculations explain the observed variation in output pump intensity and time- and energy-dependent probe intensity accurately. We showed that with accurate evaluation of all parameters, the modeling can provide detailed guidance in understanding the underlying mechanisms and in choosing materials for light propagation applications.

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